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ISOMERIZATION OF OLEFINS AND CYCLOOLEFINS IN THE PRESENCE OF GUMBRIN

Kh. I. Areshidze Submitted 17 Dec 1949

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Isomerization of olefins either by moving the double bond or by increasing the degree of branching is of great theoretical and practical significance. An isomer having the double bond nearer the center has better antiknock qualities. An increased degree of branching is also advantageous from that point of view. The development of aviation and the mechanization of the whole economy have raised the demand for fuel which is superior in quality to straight-run gasoline. The availability of cracking gasoline only partly solves the problem of satisfying the demand for liquid fuel as far as quality and quantity are concerned.

Academician N. D. Zelinskiy, Academician A. Ye. Favorskiy, Academician B. A. Kazanskiy, Corresponding Member of the Academy of Sciences of the USSR A. D. Petrov, Prof A. V. Frost, and Prof R. Ya. Levina and their collaborators have for a number of years investigated the synthesis and catalytic isomerization of olefins and cycloolefins. On the basis of this work, the improvement of cracking gasoline by catalytic treatment becomes possible and their octane numbers can be raised.

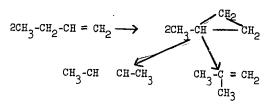
Dehydrating catalysts often have an isoermizing effect. By using clays found in the Georgian SSR (gumbrin and askanit) for the dehydration of primary and secondary alcohols, a certain amount of isomerization was obtained subsequently to the dehydration. By the dehydration of n-butyl alcohol over gumbrin, butene-1, butene-2, and 2-methylpropene-1 were obtained. In other words, there was branching in addition to the shifting of the double bond. The reaction probably proceeded as follows:

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Cyclopropane derivatives can actually be obtained by the cyclization of dehydrogenated hydrocarbons (1). The reaction scheme given above could be confirmed by proving experimentally that under different temperature conditions the conversion of 2-methylpropene-1 into butene-1 and butene-2 takes place. The reaction products were identified by bromination and by analysis of the gas according to Prof A. F. Dobryanskiy (2). 2-methylpropene-1 could be absorbed by 63 percent sulfuric acid while butene-1 and butene-2 were absorbed by 83 percent sulfuric acid. Ample data on the catalytic isomerization have been published by other investigators. Thus, 2, 6-dimethyloctene-7 was isomerized into 2, 6dimethyloctene-6 (3). Heptene-1, 3-methylhexene-2, and octene-1 could be isomerized at 450 degrees over beryllium oxide, octene-1 forming methyl heptenes (4). Displacement of the double bond occurs in the case of octene-1 as well as branching (5). The same applies to n-hexene and n-heptene (6). The optimum conditions for the isomerization of n-hexene and n-heptene were established in a series of experiments carried out at both atmospheric and elevated pressure using eitner zinc chloride or phosphoric acid as catalysts (7). The conversion of hexylene over active clay led to the formation of a mixture of n-hexane, 2-methylheptane, and 3-methylpentane (8). By the dehydration of normal primary octyl alcohol over active clay, octylene, octene, higher olefins, and saturated hydrocarbons were obtained. The reaction was accompanied by polymerization and partial hydrogenation of the olefins (9). On treatment of cracking gasoline with Caucasian clay, a sharp drop in the degree of unsaturation could be observed (10). The reaction n-butylene isobutylene was found to take place over active clay (11). Using Oglaklinsk bentonite, butene-1, could be isomerized into butene-2 (25). Redistribution of hydrogen in octene-1 and 2-ethylhexene-1 prepared at 350 degrees over aluminium oxide from the corresponding alcohols was studied. The redistribution was carried out at 250 degrees and was found to be accompanied by isomerization of the carbon skeleton (12). Patent literature contains references to the conversion of butene-1 into butene-2 under the action of phosphoric, boric, or silicic acid. C. Matignon, H. Mouren, and M. Dodé dehydrated n-butyl alcohol over aluminium oxide and obtained both butene-1 and butene-2, the latter forming from the first at 330-380 degrees due to the action of the catalyst (13). They found that high temperatures favor the shift of the double bond.

Doubts were expressed as to whether the identification of products by means of bromination and examination of the bromo-derivatives constitutes an accurate method (14, 15, 16, 17, and 18). The author of the present article regards it as accurate under the conditions used by him. He proposes to continue the investigation in order to determine the optimal conditions for the isomerization of ethylenic hydrocarbons over gumbrin and askanit (ascangel).

In the presence of gumbrin, cyclohexene could be converted into methylcyclopentenes and ethylcyclopentene into methylcyclohexyne. It was established that gumbrin is capable of both reducing and expanding cycloolefin rings.

This report was presented by the author at an out-of-town session of the Department of Chemical Sciences of the Academy of Sciences USSR held together with a meeting of the Academy of Sciences of the Georgian SSR at Tbilisi on 28 - 31 October 1949. The work in question was carried out under participation of K. Tavartkiladze at the Laboratory of Catalytic Hydrocarbon Conversion of the Institute of Chemistry of the Academy of Sciences of the Georgian SSR at Tbilisi and at the Laboratory of Organic Chemistry of the Tbilisi State University imeni I. V. Stalin.

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